Finite-size scaling in thin Fe/Ir(100) layers

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The critical temperature of thin Fe layers on Ir(100) is measured through Mößbauer spectroscopy as a function of the layer thickness. From a phenomenological finite-size scaling analysis, we find an effective shift exponent $\lambda = 3.15 \pm 0.15$, which is twice as large as the value expected from the conventional finite-size scaling prediction $\lambda = 1/\nu$, where ν is the correlation length critical exponent. Taking corrections to finite-size scaling into account, we derive the effective shift exponent $\lambda = (1 + 2\Delta_1)/\nu$, where Δ_1 describes the leading corrections to scaling. For the $3D$ Heisenberg universality class, this leads to $\lambda = 3.0 \pm 0.1$, in agreement with the experimental data. Earlier data by Ambrose and Chien on the effective shift exponent in CoO films are also explained.

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The study of finite-size effects in critical phenomena has for a long time been an issue for theoretical studies, see Refs. 1–3 for reviews. Only recently, due to important advances in epitaxial techniques necessary for the precise preparation of thin films, has this area become accessible to experiments.[4] Presently, experiments mainly study the finite-size shift of the critical temperature $T_c(n)$ of a thin film of n layers, as phenomenologically described by the shift exponent λ.

Practically, two ways of defining the shift exponent have been used. Traditionally[1], one measures the shift of $T_c(n)$ with respect to the bulk critical temperature $T_{c,b} = T_c(\infty)$ and sets

$$\delta T := \left(T_c(\infty) - T_c(n)\right) / T_c(\infty) \sim n^{-\lambda} \qquad (1)$$

in the limit when $n \to \infty$. This defines the exponent λ. Standard finite-size scaling theory relates this to the correlation length exponent ν, viz. $\lambda = 1/\nu$ (Refs. 1–3). Alternatively, as advocated for example in Ref. 5, one may also define

$$\Delta T := \left(T_c(\infty) - T_c(n)\right) / T_c(n) \sim n^{-\lambda'} \qquad (2)$$

which defines the exponent λ'. Again, the limit $n \to \infty$ is implied and from standard finite-size scaling[1–3] it follows that $\lambda' = 1/\nu$. Empirically, using ΔT rather than δT appears advantageous, since measured values for $T_c(n)$ coincide with a power law for a larger range of values of n for ΔT than for δT.

Shift exponents have been extracted successfully from either (1) or (2) for a variety of systems.[5–11] For a magnetic material adsorbed as a thin film on some non-magnetic substrate, the interaction of the magnetic moments with the substrate may restrict the degrees of freedom of the microscopic magnetic moments. Examples of thin films of magnetic materials or liquid helium realizing the $3D$ universality classes of the Ising model[6,7], the XY model[8] and the Heisenberg model[5,9], as well as the $2D$ Ising model,[10] have been constructed and analysed. The values of the shift exponent agree with those expected from $\lambda = \lambda' = 1/\nu$ to within a few per cent in all these cases, well within the experimental error bars. In practice, this is an important alternative to measure true critical exponents, since the critical region in finite-size scaling studies is much larger than in thermal measurements of the bulk quantities, as is well known.[1–3]

However, the use of (2) has been criticized in Ref. 7, on the grounds that measurements on the Néel temperatures in CoO/SiO_2 multilayers yielded[7] $\lambda = 1.6 \pm 0.1$, but $\lambda' = 3.4 \pm 0.3$, which are clearly different. It was concluded in Ref. 7 that use of eq. (2) were best avoided since the results for λ' had no interpretation.

In this letter, we shall re-examine this point and shall show how corrections to finite-size scaling may be invoked to provide a simple interpretation of those cases where a large value of λ' is measured. At the same time, we shall report data on the critical temperature $T_c(n)$ as obtained from Mößbauer spectroscopy of thin layers of Fe on Ir(100), which can be analysed and understood in the same way. As a bonus, the correction-to-scaling exponent Δ_1 can be measured experimentally.

We begin describing the experimental procedure. As Mößbauer spectroscopy is not sufficiently sensitive to get some signal on so thin layers, Fe/Ir superlayers were prepared, where the Fe/Ir bilayer is repeated 20 times. In this study, the Fe thickness was varied from 2 to 8 atomic planes and the Ir thickness is kept constant and equal to 1.5nm from one superlattice to another. The Fe/Ir(100) superlattices were prepared by molecular beam epitaxy (MBE). All the details of the sample preparation are given in Ref. 12. The Fe/Ir system is a good candidate to investigate the magnetic properties of ultrathin films, since the Fe on Ir *and* the Ir on Fe growth mode[13] is an atomic layer-by-layer growth process – also called $2D$ growth. Indeed, abrupt and flat Fe/Ir and Ir/Fe interfaces are obtained as shown by high resolution microscopy.[14] Moreover, the occurrence of $2D$ growth allows us to control accurately the atomic quantity nec-

essary to complete one atomic layer by using electron diffraction. Owing to this particular behaviour, we really deal with iron slabs constituted of exactly n atomic planes. A correct analysis of the variation of Fe magnetic properties with the number n of deposited atomic planes is thus possible. In this connection, Mößbauer spectrometry appears well suited. This technique provides a measurement of the so-called hyperfine interactions which occur between a resonant nucleus and its electronic surrounding. Among these, the magnetic hyperfine interactions manifests itself by a Zeeman splitting of the nuclear spin state when a steady local magnetic field acts on a resonant nucleus. In case of the ^{57}Fe in ferromagnetic iron, the magnetic hyperfine field arises mainly from the core polarization due to the $3d$ moment. The net $s-$spin density at the nucleus is proportional, but opposite, to the atomic on-site magnetic moment $\vec{M}$. So, the room temperature Mößbauer spectrum of standard bcc iron consists of six line patterns; the measured energy positions provide the magnitude of the hyperfine field $Bhf = 33$ T. Above the magnetic ordering temperature, due to fast thermal atomic spin flip in the paramagnetic regime, $Bhf = 0$: the splitting vanishes and the spectrum exhibits only one line (see for example Ref. 15). Usually, in case of normal bulk iron ferromagnets, the transition takes place within a narrow temperature range and the thermal variation $Bhf = Bhf(T)$ goes along with $M(T)$.[16]

As only 2% of ^{57}Fe is present in natural Fe, the MBE chamber was equipped with a 92% enriched ^{57}Fe source, in order to get a sufficient amount of ^{57}Fe in the Fe layers of the superlattices. Mößbauer spectrometry was performed in backscattered mode by detecting conversion electron after resonant absorption of 14.4 keV γ rays emitted by a ^{57}Co source. Source drive and data storage were conventional, the low-temperature electron detector was a circular microchannel plate housed in a home-made cryostat[17]. For all investigated Fe/Ir(100) superlattices, the weakening and vanishing of the Zeeman splitting was observed to take place in a rather wide temperature range, compared to the collapse which occurs for standard bcc iron when heating up. Thus, ordering temperatures of the superlattices are here defined as the temperature for which the *onset* of a line broadening takes place in the paramagnetic spectrum when cooling down from room temperature. Such a determination is illustrated in figure 1 with typical Mößbauer spectra recorded above and below the critical point. We have checked by neutron diffraction that, at least for the Fe thickness range where the Fe is still strained by Ir, there is no coupling of two Fe layers mediated the intervening Ir layer. The measured $T_c(n)$ are thus the critical temperatures of a single Fe layer. For each value of n, a single specimen was made. The quoted errors in $T_c(n)$ correspond to the steps in temperature used in the scanning of the Mößbauer spectra. For $n = 2, \ldots, 6$ deposited iron monolayers, the ordering temperatures are given in table I. For 8 monolayers, the spectrum exhibits splitting at room temperature.

The dependence of the shift $\Delta T(n)$ on n, calculated using the value $T_{c,b} = 1043$K for the critical point of bulk iron, is shown in figure 2. From at least $n = 3$ monolayers on, the data are very well described in terms of a power law.[18] Independently of any further theoretical interpretation, our data show that the system does lie inside the finite-size scaling region. Therefore, the analysis of the systematic variation of the critical point $T_c(n)$ with the number of monolayers n in terms of a phenomenological shift exponent is justified. We find

$$\lambda'_{\rm eff} = 3.15 \pm 0.15 \tag{3}$$

This is about twice the value expected from $\lambda' = 1/\nu$ in the $3D$ Heisenberg model, see table II. Since in the present setting, we explore the transition from a $3D$ bulk system to a $2D$ film, we expect that this cross-over due to finite-size effects should be described in terms of the exponents of the $3D$ universality classes.

We now come back to the problem of explaining the value of λ' found. Our starting point is the theory of finite-size scaling.[1–3] Given that the values of $T_c(n)$ (for the values of n accessible to experiment) are much lower than the bulk $T_{c,b}$, it appears sensible to consider not only the leading effects of finite-size scaling but to take finite-size corrections into account as well. For the magnetization, measured on a film of a thickness of n monolayers, we expect[1–3]

$$M_n \simeq n^{-\beta/\nu} Z\left(t n^{1/\nu}, u n^{y_3}\right) \tag{4}$$

where $t \sim T_c(n) - T_{c,b}$, β and ν are the conventional magnetization and correlation length exponents, u stands for an irrelevant scaling variable which parametrizes the leading finite-size corrections and $y_3 < 0$ is the associated exponent. Finally, $Z = Z(z_1, z_2)$ is a scaling function, which is independent of the layer thickness n.

The pseudocritical point $T_c(n)$ (and thus an associated t_n as well) is determined from the vanishing of the magnetization, viz. $M_n(t_n) = 0$. This implies

$$Z\left(t_n n^{1/\nu}, u n^{y_3}\right) = 0 \tag{5}$$

From this, we have to derive the scaling of t_n as a function of n. In particular, from figure 2 we expect that phenomenologically

$$t_n \sim \tau\, n^{-\lambda} \tag{6}$$

where $\lambda = \lambda_{\rm eff}$ is the effective shift exponent and τ is a constant. (For the purpose of this discussion, the distinction between λ and λ' as defined in eqs. (1,2) is unnecessary.) When finite-size effects are small, we can simply set $u = 0$ in (5) and then recover the standard result[1–3] $\lambda = \lambda' = 1/\nu$. Here, we want to discuss how large finite-size corrections might affect the value of $\lambda_{\rm eff}$.

For $n - 1$ monolayers, we can find, in the same way, t_{n-1} from $Z(t_{n-1}(n-1)^{1/\nu}, u(n-1)^{y_3}) = 0$. For n large enough, we can expand the arguments of Z and get, to leading order in $1/n$

$$Z\left(t_n n^{1/\nu}, u n^{y_3}\right) - y_3 u n^{y_3-1} Z_2 + \left(-\frac{1}{\nu} t_n n^{1/\nu-1} + (t_{n-1} - t_n)\, n^{1/\nu}\right) Z_1 \simeq 0 \qquad (7)$$

where $Z_{1,2} = \partial Z / \partial z_{1,2}(t_n n^{1/\nu}, u n^{y_3})$. To leading order in $1/n$, they can be considered as constants. The first term in (7) vanishes due to the condition (5), used to determine t_n. In addition, from (6) we have $t_{n-1} - t_n \simeq -\lambda \tau n^{-\lambda-1}$. Inserting into (7), we find

$$\lambda = \frac{1}{\nu} - y_3 \ , \quad \tau = -\frac{Z_2}{Z_1} \frac{y_3}{1/\nu + \lambda} u \qquad (8)$$

and, recalling that $y_3 < 0$, it is already clear that the effective shift exponent may have values larger than the usually expected $1/\nu$. The same result for λ had been obtained previously from an analysis of the finite-size scaling of the correlation length.[19]

The preceding discussion can be generalized to show that there is a whole series of correction terms to finite-size scaling,[19,20] each of them with its proper value for the effective shift exponent $\lambda_{\rm eff} = 1/\nu - k y_3$, where $k = 0, 1, 2, \ldots$. It depends on the (system-dependent and non-universal) value of u whether the leading correction is enough to describe the data or if several correction terms must be taken into account. For rather thick films, finite-size effects should be unimportant and thus $k = 0$, which reproduces the standard result $\lambda = 1/\nu$. For thinner films, one might find a cross-over into a regime described by a non-zero value of k, where $k = 1$ corresponds to the first-order calculation given above. Higher values of k apply when the lower order terms vanish.[21]

Concerning the value of y_3, we now relate it to existing theoretical predictions[22] for the three-dimensional $O(n)$ model. In these models, corrections to scaling are described in terms of the exponent Δ_1, viz. $M(t) \sim t^\beta (1 + a t^{\Delta_1} + \ldots)$, where a is a constant. Comparing with the finite-size scaling form eq. (4), then formally leads to $|y_3| = \Delta_1/\nu$. The scaling operator ψ, which generates these leading-order corrections to scaling, is well understood in a field theory setting of the $O(n)$ model. In particular, it is known[22] that ψ is *even* under spin reversal. On the other hand, the scaling operator σ which corresponds to the magnetization is *odd* under spin reversal. The amplitude τ of the first-order term calculated in (8) is proportional to the expectation value $\langle \psi \sigma \rangle_0$, to be evaluated *at* the critical point. By symmetry, this quantity vanishes. Thus, phenomenologically, we expect the finite-size data of the magnetization to be determined by the second-order corrections, with $k = 2$. We therefore arrive at the prediction, expected to be valid in cases where finite-size corrections are important

$$\lambda' = \lambda_{\rm eff} = \frac{1}{\nu}(1 + 2\Delta_1) \qquad (9)$$

In table II, we collect the field-theoretical predictions for the exponents Δ_1, $1/\nu$ and $(1 + 2\Delta_1)/\nu$. The values quoted are the mean values of those compiled in Ref. 22 from ε-expansion and from the resummed perturbation series for the three-dimensional $O(n)$ model.

We now compare (9) to the experimental data. First, consider the case of Fe/Ir(100). We expect that the ferromagnetic transition of Fe is within the Heisenberg universality class. Our result, given in eq. (3), is in good agreement with the expected value $\lambda' = 3.0 \pm 0.1$. Second, we consider the CoO/SiO_2 system.[7] CoO is known to be an antiferromagnet with localized moments and the transition is expected to be in the Ising universality class. The experimental result, obtained using eq. (2), $\lambda_{\rm eff} = 3.4 \pm 0.3$, is in agreement with the expectation $\lambda' = 3.2 \pm 0.1$ from table II. Our prediction (9) is thus clearly confirmed for two distinct universality classes. To our best knowledge, this is the first time that the correction-to-scaling exponent Δ_1 has been measured experimentally. Accepting the theoretical values of ν, we find $\Delta_1 \simeq 0.57(9)$ and $0.61(6)$ for the Ising and Heisenberg universality classes, respectively. Our findings are well consistent with the theoretical predictions obtained from field theory, see table II.

Some more comments are in order. The consistency of the measured shift exponents with $\lambda = \lambda' = 1/\nu$ in most of the systems studied so far[5–11] indicates that, usually, finite-size corrections are apparently not very important. On the other hand, in the CoO system,[7] the shift exponent had been measured using *both* δT and ΔT. While in the second case, we have shown that finite-size corrections are needed for the proper interpretation of the value of λ', the first case yields a value $\lambda = 1.6 \pm 0.1$, close to the expected $1/\nu$. We stress that it depends on the non-universal value of the coupling u (which cannot be predicted from our purely phenomenological analysis), whether a clear power law can be observed for one or other of the shift eqs. (1,2), if any, and in what regime of effective exponents the data will finally fall.

In conclusion, we have presented data on the critical temperature of thin Fe/Ir(100) layers. The thickness-dependence of $T_c(n)$ has been analysed using the phenomenological theory of finite-size scaling. Finite-size corrections were shown to play an important role in the interpretation of the effective exponent value $\lambda_{\rm eff}$, leading to the prediction (9), resolving an argument on the proper extraction of critical exponents from experimental data. Our data for Fe/Ir(100) and data[7] on CoO/SiO_2 are in agreement with the theoretical predictions coming from the $O(n)$ model. This has allowed to experimentally confirm the field-theoretic prediction of the value of the correction-to-scaling exponent Δ_1 for the $3D$ Ising and Heisenberg universality classes.

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[21] For example, if in (7) $Z_2 = 0$, one has to go to the second order terms in z_2. Phenomenologically, this leads to the same result for λ as before, with y_3 replaced by $2y_3$.
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n	2	3	4	5	6
T_c	15 ± 3	30 ± 5	70 ± 10	145 ± 15	210 ± 10

TABLE I. Transition temperatures $T_c(n)$ in Kelvin obtained from the onset of line broadening in the paramagnetic spectrum when cooling the system, as a function of the number n of Fe monolayers.

model	n	Δ_1	$1/\nu$	$(1+2\Delta_1)/\nu$
Ising	1	0.50(2)	1.586(4)	3.17(6)
XY	2	0.53(2)	1.492(7)	3.07(11)
Heisenberg	3	0.555(25)	1.413(9)	2.98(13)

TABLE II. Field theory predictions[22] from the three-dimensional $O(n)$ vector model of some critical exponents. The numbers in brackets estimate the error on the last given digits.

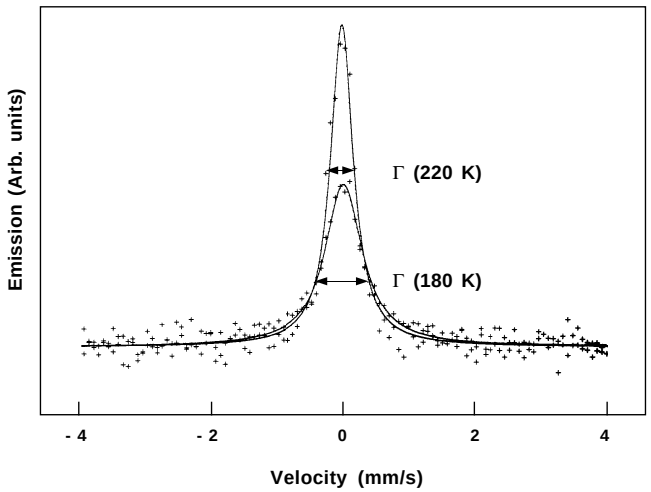

FIG. 1. Example of the Mößbauer line broadening when the temperature is decreased, for a 6ML ^{57}Fe superlattice. Down to $T = T_c$, the FWHM Γ is constant. When T_c is reached, Γ begins to increase.

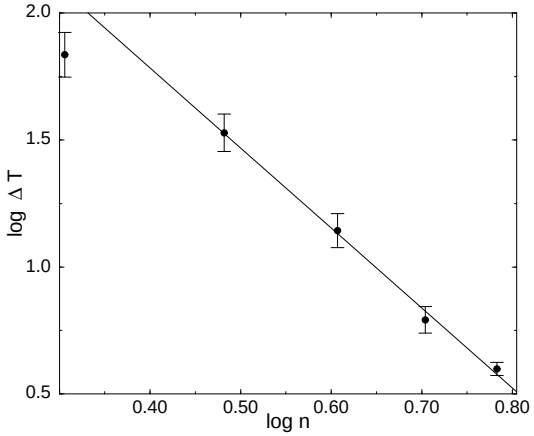

FIG. 2. Shift ΔT from eq. (2) of the critical temperature of the Fe/Ir(100) system as a function of the number n of Fe monolayers, for $n = 2, \ldots, 6$. The curve is the power-law fit $\Delta T(n) = 1069\, n^{-3.15}$.